

**REMARKS**

**Prior Rejections Withdrawn**

Applicant notes for the record that the Office has in the current Office action withdrawn the prior rejection of claims 1, 2, 22-25, 27, 28 and 51 under 35 U.S.C. 102(b) as anticipated by U.S. Pub. No. 2001/0018402 (Ostgard et al.) and the rejections of claims 3-26 and 29-87 under 35 U.S.C. 103(a) as obvious over the teachings of Ostgard et al. combined with one or more of U.S. Patent No. 6,077,620 (Pettit), "Raney Nickel-Copper Catalysts II. Surface and Pore Structures" (Wainwright et al.), U.S. Patent No. 4,820,594 (Sugita et al.) and U.S. Patent No. 6,541,142 (Yu et al.) as set forth in the Office action mailed September 7, 2006.

**Claim Interpretation**

The Office asserts on pages 2-3 of the Office action that the product-by-process limitations of claims 1, 4-6, 9-11, 33-35, 39-41, 88-90, 101, 106-108, 117-119 and 127 are not to be given any patentable weight. The case authorities cited by the Office in support of this proposition involve product-by-process claims. However, applicant is not seeking protection for the reforming catalyst *per se* defined in terms of the process by which the catalyst is prepared. The pending claims are not product-by-process claims, but are drawn to processes for reforming alcohols, wherein the reforming catalyst utilized in the process is defined in part by the manner in which the catalyst is prepared.

Moreover, applicant respectfully submits that the reforming catalyst prepared in the manner as called for in the pending claims, particularly as recited in claims 1, 88-90, 101, 106-108, 117-119 and 127, is distinguished from catalysts produced

by other methods and that these structural differences are of patentable weight, particularly in the claimed context of **using the catalyst in processes for reforming an alcohol** as defined in the pending claims such that the claimed subject matter is both novel and nonobvious over the prior art. MPEP § 2113 ("The structure implied by the process steps should be considered when assessing the patentability of product-by-process claims over the prior art. . . "). In any event, as set forth below, applicant submits that the alcohol reforming processes defined in the pending claims are patentable over the cited references independent of the manner by which the reforming catalyst is prepared as specified in the pending claims.

Contrary to the assertion on page 3 of the Office action, claims 4-6, 9-11, 33-35 and 39-41 are not drawn to the method by which the surface area is measured. Rather, these dependent claims more particularly define the claimed alcohol reforming processes as recited in the independent claims by specifying surface area characteristics of the reforming catalyst used in the process as determined by the well-known Brunauer-Emmett-Teller (BET) method. Since different surface area measuring techniques may result in different surface area determinations, it is appropriate for these claims to specify the method by which surface area is determined in order to adequately define this feature of the catalyst used in the claimed process. Nevertheless, patentability of the dependent claims reciting surface area requirements is established by the elements of the principle independent claims.

**New Rejection under 35 U.S.C. § 103(a)**

The Examiner has now rejected each of the pending claims (1-52, 88-108 and 117-130) as obvious based on the disclosure in the article "Hydrogen from Steam Reforming of Ethanol

Characterization and Performance of Copper-Nickel Supported Catalysts" (Marino et al.) in view of U.S. Patent No. 2,892,801 (Sargent). Applicant respectfully requests reconsideration and withdrawal of the rejection of the pending claims.

Claim 1 is directed to a process for reforming an alcohol. The process comprises contacting a feed gas mixture comprising an alcohol with a reforming catalyst to produce a reforming product mixture comprising hydrogen. The reforming catalyst contacted with the feed gas mixture comprises a metal sponge supporting structure and a copper coating at least partially covering the surface of the metal sponge supporting structure wherein the metal sponge supporting structure is prepared by a process comprising leaching aluminum from an alloy comprising aluminum and a base metal (e.g., nickel, cobalt, copper).

Independent claim 27 is directed to a process for reforming ethanol. The process of claim 27 comprises contacting a feed gas mixture comprising ethanol with a reforming catalyst at a temperature below about 400°C to produce a reforming product mixture comprising hydrogen. The reforming catalyst contacted with the feed gas mixture comprises copper at the surface of a metal supporting structure.

Independent claim 117 is directed to a process for reforming an alcohol. The process comprises contacting a feed gas mixture comprising an alcohol with a reforming catalyst to produce a reforming product mixture comprising hydrogen. The reforming catalyst contacted with the reforming catalyst is prepared by a process comprising depositing copper on a metal sponge supporting structure.

It has been found in accordance with the claimed invention that catalysts comprising copper at the surface of metal supporting structures exhibit the mechanical strength, high surface area, high thermal conductivity, and desired catalytic

activity that it is particularly suited for gas-phase reforming of alcohols. In particular, the catalysts used in the practice of the present invention are stable in and particularly active for the gas-phase, thermal decomposition of ethanol into hydrogen, methane, carbon dioxide, and carbon monoxide at moderate temperature (i.e., below about 400°C).

Marino et al. report an investigation of the effects of copper loading and calcination reaction temperature on catalysts comprising copper and/or nickel supported on K-doped  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> spheres. Several catalysts with differing amounts of copper and nickel loading were characterized by their temperature-programmed-reduction (TPR) profile and x-ray diffractograms. The catalysts were also characterized by their activity and selectivity in steam reforming of ethanol.

Sargent is directed to copper-plated hydrogenation catalysts produced by reacting Raney nickel in copper salt solutions. Catalysts with various copper loadings were characterized by the activity of the catalyst toward the liquid-phase hydrogenation of glucose to sorbitol.

The Office asserts on pages 5-6 of the Office action that it would have been obvious to one skilled in the art to use the copper-plated nickel sponge of Sargent as the catalyst in the reaction of Marino et al. since Marino et al. teach that the presence of nickel improves the performance of the catalyst. Applicant submits that the assertion by the Office misconstrues the disclosure of Marino et al., particularly as to what the disclosure of Marino et al. suggests to a person skilled in the art.

Nowhere does Marino et al. suggest substitution of the **ceramic**,  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supporting structure of the catalyst described therein with any other type of supporting structure and certainly not substitution with a **metal** (e.g., a metal sponge)

supporting structure as recited in independent claims 1, 27 and 117. Furthermore, the teaching of Marino et al. that nickel may be suitably incorporated into the catalyst to promote hydrogen selectivity (last open paragraph of the second column of p. 1098; third open paragraph of the second column of p. 1100) merely suggests to one skilled in the art to incorporate nickel in a ceramic-supported copper catalysts by depositing nickel onto the **surface** of the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supporting structure (See "Catalyst Preparation", p. 1095). In fact, Marino et al. actually teach away from use of supporting structures other than  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> by emphasizing the importance of the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supporting structure in the ethanol reforming reaction. For instance, Marino et al. disclose that copper-nickel  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>-supported catalysts "present acceptable activity and selectivity" for the reforming of alcohol (third open paragraph of the first column of p. 1095), but fail to disclose suitability of any catalyst containing a supporting structure other than  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> as an alcohol reforming catalyst. In addition, Marino et al. suggest that the catalyst described therein is suitable not just because of its copper-nickel coating, but because of the interactions of the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> support with that coating ("In all samples analyzed, nickel is present as NiAl<sub>2</sub>O<sub>4</sub>"; second open paragraph of the second column of p. 1100).

More importantly, those skilled in the art of catalysis appreciate the substantial unpredictability in the development of catalytic reaction systems. In catalysis, the nature of the support is crucial. Supports play a key role in determining the stability of catalysts, since they separate the metal centers and prevent deactivation by sintering. Supports also typically influence activity and selectivity and the performance of one type of material in supporting a catalytic active phase for a

desired reaction cannot be expected to translate to a different support material for the same active phase and reaction. With that appreciation, the skilled person would not extrapolate Marino et al.'s teaching regarding possible **surface** addition of nickel to a  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>-supported copper catalyst as even remotely suggesting that substituting the ceramic support with a metal supporting structure (e.g., the nickel sponge supporting structure of Sargent) would provide a catalyst capable of replicating or similarly enhancing the steam reforming reaction kinetics demonstrated by the copper-nickel  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>-supported catalyst. Possible surface addition of nickel to a ceramic-supported copper catalyst simply does not direct one skilled in the art of catalysis to attempt substituting a metal support structure for the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> spheres used in Marino et al.

Moreover, if one skilled in the art had reason to substitute the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supporting structure of the copper-nickel reforming catalyst of Marino et al. with another support (which applicant does not admit), the skilled person would not look to the Raney nickel support of Sargent as a suitable support for use in the steam reforming of ethanol. Nothing in the disclosure of the primary reference teaches that a metallic nickel support (i.e., Ni(0)) such as that used in Sargent is capable of replicating the alcohol reforming activity of the supported NiAl<sub>2</sub>O<sub>4</sub> complex (containing Ni(II)) in the catalyst of Marino et. al. That is, the metallic nickel in the support of Sargent is not in the form of Ni(II) present in the NiAl<sub>2</sub>O<sub>4</sub> complex believed to be active in Marino et al. Further, the alcohol reforming processes of the present invention requires contacting a feed **gas** mixture comprising the alcohol to be reformed with the reforming catalyst. Similar to U.S. Publication No. 2001/0018402 (Ostgard et al.), which formed the basis of the novelty and several obviousness rejections of the

Office action mailed September 7, 2006, Sargent fails to teach or suggest that the catalyst he discloses is suitable for any **gas-phase** reaction, and particularly, the **gas-phase dehydrogenation** of an alcohol as called for in the pending claims. Rather, the reactions actually demonstrated by Sargent are all **hydrogenation** reactions and are conducted in the **liquid-phase**. While Sargent makes passing reference to suitability of the catalysts described therein for "**certain** dehydrogenation, dehalogenation and desulfurization reactions," nothing in the reference suggests to one skilled in the art that gas-phase reforming of alcohols is among the certain dehydrogenation reactions for which the catalyst might be suitable. One skilled in the art would not substitute Sargent's Raney nickel supporting structure for the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supporting structure used in the experimental ethanol reforming process of Marino et al. with a reasonable expectation of success since the process of Marino et al. is a gas-phase dehydrogenation reaction and nothing in the disclosure of Sargent directs one skilled in the art to use a metal sponge supported catalyst in a gas-phase dehydrogenation reaction.

Applicant respectfully submits that in light of the preceding arguments the Office has failed to establish a *prima facie* case of obviousness with respect to the invention as defined by independent claims 1, 27 and 117 and all claims depending therefrom. Accordingly, favorable reconsideration and allowance of all pending claims are respectfully requested.

### **Unexpected Results**

Applicant further submits that even if the Office were to establish a *prima facie* case of obviousness, the pending claims are patentable over the cited references. Evidence of unexpected advantageous properties such as superiority of a

property shared with the prior art rebuts a *prima facie* case of obviousness. MPEP § 716.02(a). Applicant submits that the process of independent claims 1, 27 and 117 are further nonobvious over the cited references as they result in superior alcohol conversion rates.

Marino et al. describe the conversion percentage of ethanol for six copper-nickel,  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>-supported catalysts with 0, 1.26, 2.17, 2.17 (calcined at 800°C rather than 550°C), 4.75 and 6.36% by weight copper loading in Table 2 (p. 1099). The reaction was carried out at 300°C and atmospheric pressure. A water-ethanol mixture of a molar ratio 2.5:1 was fed at 0.12 ml/min and was vaporized and passed over 2.5 g of the catalyst packed in a 20 mm diameter fixed bed reactor.

In Example 5 (Tables 5 and 6) of the application, applicant describes conversion of ethanol under similar reaction conditions. A 30% by volume ethanol, 70% by volume water-ethanol mixture (corresponding to about a 2.5:1 molar ratio) was passed over a copper-plated, nickel sponge supported catalyst under 30% by weight copper-loading at 275°C and with an upstream pressure less than 5 psig. The water-ethanol mixture was fed at a rate of 0.1 ml/min and was vaporized and passed over 2.5 g of the catalyst packed in a 12.7 mm diameter, 457.2 mm in length fixed-bed reactor.

As can be seen by comparing Table 2 of Marino et al. to applicant's Tables 5 and 6, applicant's process possesses superior capacity for ethanol conversion. After a run-time of 90 minutes the catalyst of Marino et al. is not capable of achieving better than 90% conversion and, rather, more typically achieves an ethanol conversion of less than 70%. In contrast, use of the copper-plated, metal supported catalyst in the practice of the process of the present invention achieves about 100% conversion up to a 265 hour run time. When the temperature



is increased from 275°C to 300°C, conversion returns to 100% for at least a 310 hour run-time.

In light of the surprisingly superior alcohol conversion and the high activity of the catalyst at lifetimes of more than 300 hours in accordance with applicant's process as compared to the process of Marino et al. and the fact that Sargent is fully silent as to the superior conversion capability in gas-phase alcohol reforming reactions of the catalyst described therein, applicant submits that the invention defined in the pending claims is nonobvious over the cited references.

Applicant authorizes the Office to charge the fee for a one month extension of time to Deposit Account No. 19-1345. Applicant authorizes the Office to charge any fee deficiency or credit any overpayment in connection with this response to Deposit Account No. 19-1345.

Respectfully submitted,

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